Fukushima-derived fission nuclides monitored around Taiwan: Free tropospheric versus boundary layer transport

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A B S T R A C T

The 2011 Fukushima nuclear accident in Japan was the worst nuclear disaster following the 1986 Chernobyl accident. Fission products (nuclides) released from the Fukushima plant site since March 12, 2011 had been detected around the northern hemisphere in about two weeks and also in the southern hemisphere about one month later. We report here detailed time series of radioiodine and radiocesium isotopes monitored in a regional network around Taiwan, including one high-mountain and three ground-level sites. Our results show several pulses of emission from a sequence of accidents in the Fukushima facility, with the more volatile 131I released preferentially over 134Cs and 137Cs at the beginning. In the middle of the time series, there was a pronounced peak of radiocesium observed in northern Taiwan, with activity concentrations of 134Cs and 137Cs far exceeding that of 131I during that episode. From the first arrival time of these fission nuclides and their spatial and temporal variations at our sampling sites and elsewhere, we suggest that Fukushima-derived radioactive materials were transported to Taiwan and its vicinity via two pathways at different altitudes. One was transported in the free troposphere by the prevailing westerly winds around the globe; the other was transported in the planetary boundary layer by the northeast monsoon wind directly toward Taiwan.

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1. Introduction

The Fukushima nuclear accident occurred in the wake of the double disaster of the 9.0 magnitude Tohoku earthquake and tsunami on March 11, 2011. The earthquake triggered the shutdown of the three active reactors at the Fukushima Daiichi nuclear power station, and the tsunami stopped the station’s backup diesel generators, causing a station blackout. The subsequent lack of electric cooling led to a series of explosions and complete melt downs of three active reactors’ cores at the Fukushima facility, with problems at all six reactor units and the central spent fuel pool (Wikipedia website). At the first several days of the accidents, air transport in the mid-latitudes was dominated by prevailing westerly winds, which could circle around the globe in 2–3 weeks (Uno et al., 2009; Warneck, 2000). Therefore, if Fukushima-derived radionuclides were introduced into and above the planetary boundary layer, their dispersal on both regional and global scales would be inevitable. Indeed, fission products released from the accidents had since been spread around the globe (information available from the website of CTBTO Preparatory Commission). For instance, it took only 4 days for the gaseous 133Xe ($T_{1/2} = 5.2$ d) to be transported >7000 km eastward, from the emission point in Fukushima Japan (37°25′17″N, 141°1′57″E) across the Pacific Ocean, and be detected on March 16 at the Pacific Northwest Laboratory in Richland, Washington, U.S.A. (46°16′47″N, 119°16′53″W) (Bower et al., 2011). In the ensuing days, 131I ($T_{1/2} = 8.02$ d), 134Cs ($T_{1/2} = 2.06$ yr) and 137Cs ($T_{1/2} = 30.1$ yr) originated from Fukushima were detected across the contiguous United States, progressively from the western seaboard to the eastern seaboard (US Environmental Protection Agency website). By March 24, Fukushima-derived radioactive materials had traveled across the Atlantic and been detected at many CTBTO (Comprehensive Nuclear-Test-Ban Treaty Organization) monitoring stations and elsewhere in Europe (Masson et al., 2011), from the Iberian Peninsula (Lozano et al., 2011) to Thessaloniki, Greece (Manolopoulos et al., 2011). As of April 13, about one month after the nuclear accident, Fukushima-derived radioactivity had spread to the southern hemisphere and had been detected in the Asia-Pacific region at stations located for example in Fiji, Malaysia, Papua New Guinea and even Australia (CTBTO Preparatory Commission website).

Compared with the information about the dispersion of Fukushima-derived fission nuclides in the western hemisphere, much less data is available in eastern Asian countries and we are not aware of any data outside CTBTO’s signatory nations. Based in Taiwan, we have been continuously collecting aerosol samples on a daily basis from a ground-level network and high-mountain sites. The samples were routinely analyzed by nondestructive gamma spectrometry for two radionuclides commonly used as aerosol tracers (i.e., 7Be and 210Pb) followed by...
were detected coincidentally on the air filters collected from late March to late April subsequent to the Fukushima nuclear accident. We feel it warranted to make a timely report of the time series and discuss its implications for radiation safety and atmospheric transport.

2. Observation sites and methods of sampling and analysis

Fig. 1 shows the location of our sampling sites with respect to that of Fukushima in eastern Japan. Of the four sampling sites, two are located in Taiwan (NK and MLL) and the other two are on offshore islets (PCY and DS). The choice of these sites for this particular study is dictated by their altitude and orientation in the regional and global wind field. MLL (Mt. Lulin; 23°28’07”N, 120°52’25”E, 2862 m) is a high-mountain site in Taiwan’s Central Range, which is under the influence of the westerlies, especially from March to May. The other three ground-level sites are confined in the boundary layer and are influenced primarily by East Asian monsoon which blows from the northeast toward the southwest in this season. NK (Nankang; 25°02’26”N, 121°36’50”E,) is in the Taipei Basin in northern Taiwan; PCY (Pengchiayu Islet; 25°37’12”N, 122°4’12”E) is upwind in the southern East China Sea; and DS (Dongsa Islet; 20°41’54”N, 116°43’43”E,) is downwind in the northern South China Sea.

The aerosol samples were collected by 24-hour pumping of ~1400–2000 m³ air through cellulose and/or glass fiber filters (8 in. × 10 in. in size). A total of five HPGe detectors with 100–150% relative efficiency (with respect to 3×3 NaI) were employed to provide the necessary throughput for counting daily samples returned from all sites in a timely manner. Absolute efficiencies of the detectors for counting nuclides on the filter samples were calibrated using blank filters spiked with a mixed reference materials containing known activity concentrations of 210Pb, 214Pb (in equilibrium with 226Ra), 134Cs and 137Cs, among other nuclides. The efficiencies for counting 134Cs and 137Cs were calculated directly and that for 131I was derived from the efficiency curve obtained by least-squares fitting. Activity concentrations of 131I, 134Cs and 137Cs in each sample were calculated based on net counts registered under the photon peaks centered at 364.48 keV, 604.66 keV and 661.62 keV, respectively. A sample spectrum is given in the Supplementary Material (Fig. S1). The minimum detection limits (MDL) of the nuclides of interest can be approximately evaluated using the equation:

$$ MDL = A \times 3\sqrt{C_{bgd}/C_{sample}} $$

where A represents the activity concentration of the nuclide of interest in the sample, $C_{sample}$ is the accumulated count in the photon peak of interest in the sample spectrum, and $C_{bgd}$ is the corresponding count from the spectrum obtained by counting blank filters for the same length of time. Since the accumulated counts ($C_{sample}$ and $C_{bgd}$) are proportional to the counting time ($T$), it can be shown that $MDL \propto 1/\sqrt{T}$. Thus, MDL is actually a dependent variable whose value can be reduced by increasing the sample size (i.e., air volume pumped through the filters) and/or the counting time. To maintain the sample throughput and achieve the specified temporal resolution, the counting time of our samples varied between 12 and 48 h, resulting in detection limits generally on the order of μBq/m³, significantly below the A values reported here for all three nuclides.

It should be noted here that, using the filtration method, the measured radioactivities represent those in the particulate form. Although radioactivity in the gaseous form may be minor or negligible for the particle-reactive 137Cs, it could constitute a major fraction of the total 131I (Masson et al., 2011; Morino et al., 2011), especially near the source of emission. However, for the purpose of this study, it is unnecessary and actually provides no advantage to measure absolute and total (i.e., gaseous plus particulate) activities, which requires the
use of charcoal cartridges. Rather, it should be more appropriate to collect large-volume samples by the filtration method so that the time of first appearance can be captured precisely and the waxing and waning of these fission nuclides’ activity concentrations in the atmosphere can be defined better.

3. Results and discussion

The time series of $^{131}$I, $^{134}$Cs and $^{137}$Cs activity concentrations are summarized in two ways. They were shown in Fig. 2 using linear scale, with all three nuclides plotted together for each individual site. To facilitate inter-site comparison, the same data are presented in Fig. 3 on logarithmic scale, with all sites plotted together for each nuclide and the $^{131}$I/$^{137}$Cs activity ratio. The entire datasets can be found in Table S1 of the Supplementary Material.

3.1. First arrival time at different sites and the speed of transport

Massive releases of radioactive materials from the Fukushima Daiichi nuclear power plant likely happened during March 12–16 due to emergent venting and hydrogen explosions in four reactor units. Consequently, radiation dose in the air around the plant peaked on March 15 (Chino et al., 2011) and stayed at the highest levels until March 16 (Wikipedia website). The modeling after the accident showed that the wind regime in the vicinity of the Fukushima facility was predominately west, transporting radioactive material predominantly to the east (CTBTO Preparatory Commission website; Takemura et al., 2011). It was not until March 17 that phases with north-easterly winds occurred. The first arrival times of Fukushima-derived fission nuclides in the southwest are March 24 at Okinawa (CTBTO Preparatory Commission website), March 25 at PCY and NK, March 28 at DS, and March 29 at MLL. We could not detect any slighter activity at PCY and NK in northern Taiwan, nuclide activity levels were similar and the rises and falls in the time series at these two sites were almost synchronous (see Fig. 3). However, since PCY is an offshore islet in the upwind direction whereas NK is downwind in the Taipei Basin surrounded by mountains, some discrepancies between these two sets of data can be expected at times. For example, before the radiation cloud entered the Taipei Basin in early April, there was episodic rain in the mountainous north. Higher $^{131}$I/$^{137}$Cs ratio at NK (~1) than at PCY (~0.3) on April 6 could be caused by preferential removal of the particle-reactive $^{137}$Cs relative to $^{131}$I before the air mass arrived at NK.

The progressive arrival of Fukushima-derived fission nuclides in the southwestern direction is supported by the model simulation (to be discussed later) and is consistent with transport via the northeastern monsoon wind at speeds estimated as follows. Based on the distance from Fukushima to PCY (~2200 km) and the transit time of Fukushima-derived fission nuclides (~8 days, from March 17 to March 25), the radioactive material was transported by the northeastern airflow at a mean speed of ~3 m/s toward the southwest. The arrival time of the much diluted radiation plume at DS, ~770 km downwind from PCY, is ~3 days later (on March 28), which also yields a mean speed of ~3 m/s. As regards the same data monitored at MLL, the activity levels are substantially lower and the pattern of the time series is entirely different from those observed at the ground-level sites mentioned above. We ascribe it to a different transport pathway at much higher altitudes. To substantiate this, we summarize in Fig. 4 the time series of $^{131}$I at stations influenced by the westerlies. Besides a systematic decrease of the initial activity concentrations from the source point eastward, the first arrival time of Fukushima-derived $^{131}$I was March 18 at Berkeley, California 37°52′18″N, 122°16′22″W (data from UCB website), March 24 at Thessaloniki, Northern Greece 40°38′1″N, 22°58′E (Manolopoulou et al., 2011) and March 29 at MLL of Taiwan. Given the approximate distance eastward around the globe from Fukushima to MLL (~32,380 km) and the transit time (17 days since March 12), we calculated a mean eastward speed of ~22 m/s for the transport of Fukushima-derived nuclides, consistent with the speed of the westerly wind in the free troposphere around the globe (Bluth et al., 1992). Upon arrival at MLL, the activity levels of $^{131}$I were two orders of magnitude lower at times. For example, before the radiation cloud entered the Taipei Basin in early April, there was episodic rain in the mountainous north. Higher $^{131}$I/$^{137}$Cs ratio at NK (~1) than at PCY (~0.3) on April 6 could be caused by preferential removal of the particle-reactive $^{137}$Cs relative to $^{131}$I before the air mass arrived at NK.

### Fig. 2. Time series of $^{131}$I, $^{134}$Cs and $^{137}$Cs activity concentrations at PCY (a), NK (b), DS (c) and MLL (d). Horizontal bars denote sampling interval (mostly 24 h) and vertical bars represent ±1σ around the mean based on counting statistics. The entire datasets can be found in Table S1 in the Supplementary Material.
lower than those observed at Berkeley, California. Although $^{131}I$ is a fairly short-lived nuclide, such a decrease cannot be explained solely by radioactive decay; it must involve mixing of the radioactive cloud with, and hence dilution by, ambient air en route. The around-the-globe journey in the northern hemisphere renders the Fukushima-derived radioactivity to be picked up later at MLL, a high-mountain site, than at DS, near the sea level, despite that the horizontal distance between Fukushima and DS ($\sim 3000$ km) is ten times shorter than that from Fukushima to MLL.

### 3.2. Temporal variation in activity concentrations and isotopic composition

Besides the timing of the first detection of Fukushima-derived fission nuclides, it is important to note the episodic signals in the time series, which are most pronounced at PCY, the upwind ground-level station. During the early stage of the nuclear accident, hardly had one pulse subsided when another rose. Conceivably, the release of fission products from the Fukushima plant must be related to a series of hydrogen explosions in the nuclear facility which ejected radioactive vapors into the atmosphere at altitudes favorable for long-distance transport. It is also informative to note the relative variation of radioiodine and radiocesium activities released during the nuclear accident. While the $^{134}Cs/^{137}Cs$ activity ratio maintained fairly constant (largely between 0.8 and 1), the $^{131}I/^{137}Cs$ activity ratio varied substantially in the course of the accident. At the initial stage of the time series $^{131}I/^{137}Cs$ ratios were substantially higher than those monitored later, indicating preferential release of the more volatile $^{131}I$ at the early phase of the accident. At PCY, for example, the ratio dropped from >20 during March 26–27 to ~0.3 during April 6–7 (Fig. 3d). Such a decrease cannot be ascribed to decay of $^{131}I$ because the second pulse of $^{131}I$ in early April was even stronger and lasted longer than the first one in late March (Fig. 2a). In fact, the $^{131}I/^{137}Cs$ activity ratio monitored in late April (~1) was significantly higher than that during April 6–7 (~0.3). The lowest $^{131}I/^{137}Cs$ activity ratio is associated with a pronounced radiocesium peak during April 6–7 which is clearly decoupled from those major $^{131}I$ pulses. The temporal trend of the $^{131}I/^{137}Cs$ ratio we observed in northern Taiwan is fairly similar to that monitored near the Fukushima site in showing higher ratios in the early part of the time series, with a fluctuation up to two orders of magnitude for daily measurements in a

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**Fig. 3.** Inter-site comparison of the time series of $^{131}I$ (a), $^{137}Cs$ (b), $^{134}Cs$ (c) and the $^{131}I/^{137}Cs$ activity ratio (d).

**Fig. 4.** A comparison of $^{131}I$ monitored at three stations located in the westerlies. The Berkeley, California and northern Greece time series are available from the website http://www.nuc.berkeley.edu/AirSamplingResults and Manolopoulou et al. (2011), respectively.
month-long period (Chino et al., 2011). As mentioned earlier, this ratio may be affected by fractionation during rainfall. It also bears information about nuclear reactions in the damaged reactors and when these fission products were released (Matsui, 2011).

3.3. A model simulation of boundary layer transport

The WRF/Chem (Ver. 3.1) tracer model (Grell et al., 2005) was employed to verify the southward transport of radioactive materials from Fukushima to Taiwan. To run the model, we adopted the Yonsei University (YSU) planetary boundary layer scheme (Hong and Dudhia, 2003; Lin et al., 2009), with the meteorological initial and boundary conditions obtained from NCEP-GFS data sets at 3-hour intervals. The horizontal resolution for the simulation was 20 km and the grid box has 400 x 400 points in the east–west and south–north directions. There are 35 layers with the lowest level about 20 m above the surface. To assure the meteorological fields were well simulated, the four-dimensional data assimilation (FDDA) scheme was activated based on the NCEP-GFS analysis data. The tracers in the model were assigned to the grid of the Fukushima site and the variable release rates were based on estimates made by Chino et al. (2011), which decreased from values as high as 10^7 TBq/h on March 15 to 1 TBq/h during early April. Shown in Fig. 5 is a glimpse of the model simulation (please refer to Animation 1 in the Supplementary Material for the details). It is consistent with CTBTO’s simulation (CTBTO website) in showing phases of north-easterly winds in the first week of April, 2011, transporting Fukushima-derived radionuclides predominately toward the southwest, with radioactivity peaked during April 6–7, 2011. The model simulation is well supported by our time series data at PCY and NK (Fig. 2a and b), which clearly show the radiocesium peak and the highest total radioactivities (i.e., ^131I + ^134Cs + ^137Cs) during April 6–7 (see Fig. S1).

3.4. Some implications of the observation and modeling results

Because of reactor meltdowns, the Fukushima nuclear accident was rated at the highest INES (International Nuclear and Radiological Event Scale) level of 7, similar to that rated for the Chernobyl accident 25 yr ago. However, there are fundamental differences between these two accidents. At Fukushima, the peak radiation level observed as of March 16, 2011 was 1000 mSv/h near the reactor core, much lower than levels of 10,000 mSv/h recorded outside the reactor building in Chernobyl (Wikipedia website). In the Chernobyl event, the reactor core itself exploded, which had the effect of spreading various kinds of fission products away from the accident center. In the Fukushima event, the lack of refractory fission products such as ^95Zr, ^140Ba, and ^140La detected outside Japan probably suggests less severe environmental impact compared with that caused by the Chernobyl accident. However, the total amount of radioactivity released from the Fukushima plant into the environment is highly uncertain (Chino et al., 2011; Morino et al., 2011; Stohl et al., 2011; Yasunari et al., 2011) and is complicated by releases both into the atmosphere and ocean waters off Japan. Outside Japan, Fukushima-derived fission nuclides were first detected in the air and rainwater samples collected in the west coast of the United States where the measured activity levels and their equivalent radiation doses did not pose any health risks to the public (US EPA website, Norman et al., 2011). Likewise, the accident was not expected to cause health concerns in Europe (Masson et al., 2011) and in Asia except Japan. Airborne activity levels of both radioiodine and radiocesium isotopes around Taiwan were among the lowest detected worldwide, and they had declined rapidly from the peak values detected in early April. Since early May, 2011, activities of these nuclides in samples returned from our monitoring sites have fallen below our detection limits.

Taiwan is ideally located in the western Pacific to study atmospheric transport between and across the largest continent and the
largest ocean on Earth. The distribution of our monitoring sites allows us to collect aerosols of various kinds and study their sources and transport pathways. Taking advantage of our sampling network, we have been monitoring the transport of eolian dust from the Asian continent and beyond toward the Pacific. It is pertinent to point out here that, during March 2010, exactly one year before the Fukushima event, a 20-year super dust storm originated from north China hit the western Pacific. Signals of that dust storm were unequivocally recorded at PCY, NK and DS, but not at MLL, suggesting transport in the planetary boundary layer through regional weather systems. From back-trajectory analysis, satellite remote sensing and model simulation, eolian dust collected at MLL can be traced to non-Asian sources including North Africa and Middle East, suggesting an integrated source from the global dust belt and transport by the westerly wind in the free troposphere (Hsu et al., unpublished results). One year after, using Fukushima-derived nuclides as tracers, we have made basically the same findings about atmospheric transport. We believe our data will contribute to the database for Fukushima-related studies and for testing climate models in the future.

Supplementary materials related to this article can be found online at doi:10.1016/j.epsl.2011.12.004.

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